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<p>A flow-drift tube instrument has been employed to measure the mobilities of five cluster ion systems that have been detected in the earth's atmosphere. The mobilities of $\text{NO}^+(\text{H}_2\text{O})_{0-2}$, $\text{H}_3\text{O}^+(\text{H}_2\text{O})_{0-3}$, $\text{NH}_4^+(\text{NH}_3)_{0-3}$, $\text{NH}_4^+(\text{CH}_3\text{CN})_{0-3}$, and $\text{NO}^+(\text{CH}_3\text{CN})_{0-3}$ were determined as a function of field strength by the dual ion signal depletion method. For $\text{H}_3\text{O}^+(\text{H}_2\text{O})_{0-3}$ cluster ions, the reduced zero-field mobility decreases by almost equal amounts as a function of n, whereas for the $\text{NH}_4^+(\text{NH}_3)_{0-3}$, $\text{NH}_4^+(\text{CH}_3\text{CN})_{0-3}$, and $\text{NO}^+(\text{CH}_3\text{CN})_{0-3}$ cluster ions, the decrease in mobility with cluster size is more gradual for higher n. A simple geometrical model based on the effective cross-sectional area of the cluster ions is consistent with the observed n-dependence of the mobilities.</p>			
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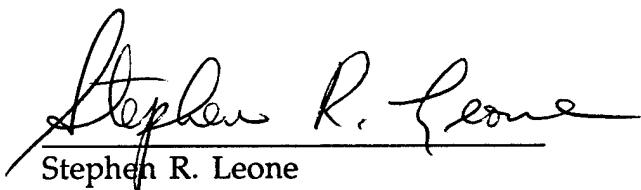
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Stephen R. Leone

"REACTIONS OF ATMOSPHERIC CLUSTER IONS"

Annual Technical Report

June 1, 1994 - May 31, 1995

Cluster ions play an important role in the ion chemistry of the earth's atmosphere. For example, clusters with NO^+ , H_3O^+ and NH_4^+ as core ions, and with H_2O , NH_3 or CH_3CN as solvating ligands, among many others, have been detected in the earth's troposphere, stratosphere and ionosphere. Although the association and dissociation processes forming these ions are in general well-understood, their mobilities are largely unknown. These values are essential, not only in modeling atmospheric phenomena, but also in providing direct information about the ion-buffer gas interaction potential.

Our selected ion flow tube apparatus has been modified to include a well-defined flow drift region and instrumentation for ion modulation and data acquisition. Core ions are generated in an ion source, mass-selected and injected into the flow tube where they associate with added solvent molecules before entering the drift region. Two drift rings, at known separation, are simultaneously pulsed; the resulting ion depletions are detected with a quadrupole mass filter in a time-resolved manner, as a function of E/N. The measured zero-field mobilities for five cluster systems in helium are summarized in Table 1.

Table 1. Reduced zero-field mobilities ($\text{cm}^2 \text{V}^{-1} \text{s}^{-1}$) of cluster ions at 300K

$\text{A}^+(\text{B})_n$	n=0	n=1	n=2	n=3
$\text{NO}^+(\text{H}_2\text{O})_n$	22.4 ± 0.5	16.8 ± 0.5	12.9 ± 0.3	
$\text{H}_3\text{O}^+(\text{H}_2\text{O})_n$	21.5 ± 0.5	17.6 ± 0.4	13.7 ± 0.3	10.4 ± 0.3
$\text{NH}_4^+(\text{NH}_3)_n$	22.1 ± 0.6	15.7 ± 0.3	11.4 ± 0.3	10.2 ± 0.3
$\text{NH}_4^+(\text{CH}_3\text{CN})_n$	22.1 ± 0.6	12.3 ± 0.5	6.8 ± 0.2	6.7 ± 0.2
$\text{NO}^+(\text{CH}_3\text{CN})_n$	22.4 ± 0.5	12.3 ± 0.3	7.9 ± 0.5	7.9 ± 0.6

The observed size dependence of the cluster ion mobilities can be divided into two classes. For $\text{H}_3\text{O}^+(\text{H}_2\text{O})_n$ with n=0-3, the mobilities decrease uniformly as the cluster size increases; this behavior reflects the systematic increase in the ionic cross section as water molecules add to the planar ionic structure. In contrast, for three other systems, the mobility decreases less dramatically for higher n species, and ions with two and three solvent molecules have similar mobilities. This behavior reflects a "tetrahedral" geometry where the effective ionic cross section increases only slightly during the filling of the first solvation shell. These experimental findings are consistent with simple molecular modeling. We are currently studying the mobilities of clusters with other core ions and solvent molecules, mixed cluster systems and the effect of other buffer gases.

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